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Title:	Exploring Neutron Imaging for Non-DE Analysis of corrosion in ICCWR of 3013 Containers
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Exploring Neutron Imaging for Non-DE Analysis of corrosion in ICCWR of 3013 Containers

Acknowledgments:

MIS team (AMPP4): Juan G. Duque, Daniel Rios, Joshua E. Narlesky, Ed Romero,
Mary Ann Stroud, Douglas Veirs, Elizabeth Kelly, Kim Kaufeld, Laura Worl, John Berg (C-PCS)
Erik B. Watkins (MPA-11), Markus Strobl and Jacopo Valescchi (PSI), Switzerland, El’ad Caspi (NRCN), Israel

Packaging style

DOE stores excess weapons plutonium in nested, welded stainless steel containers (3013 Containers)

3013 containers: 3 layers

1. Convenience container (different features depending on origin, some have filters for outgassing, ie: Hanford-Livermore)-CC. In contact with material.

2. Inner container (welded)-IC. In contact with gases generated by the material.

3. Outer container (welded)-OC.

Expected lifetime of packaged 3013 containers is 50 years.

Corrosion Observations at DE

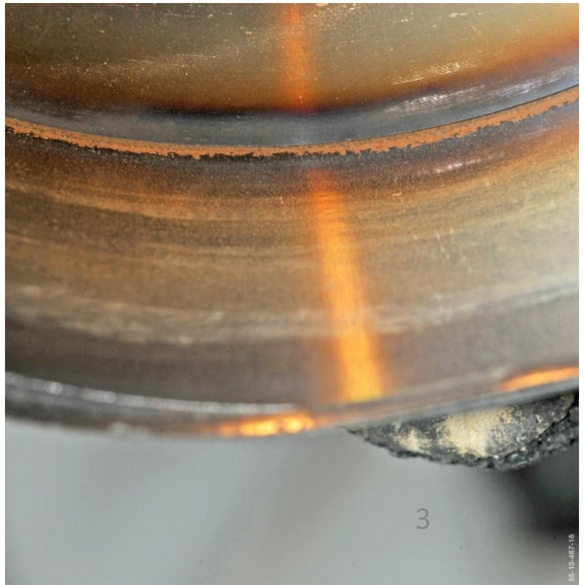
DE of some 3013 container packaged with chloride-bearing PuO_2 and moisture have shown corrosion on the convenience and inner containers.

Small scale corrosion experiments suggest chlorine-containing gases (HCl and Cl_2) may have caused the observed headspace corrosion in 3013 containers.

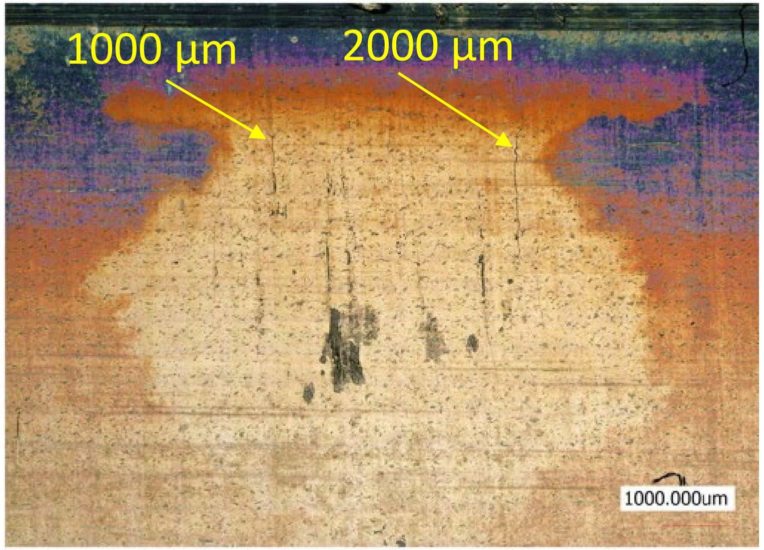
- $CaCl_2$ supports release of Cl_2 and HCl
- $MgCl_2$ supports release of HCl only

Convenience Can

Inner Can Lid



Most Common Corrosion Analysis Techniques used in 3013 containers



Laser confocal microscope (LCM)

Identification of pits, cracks and surface corrosion.

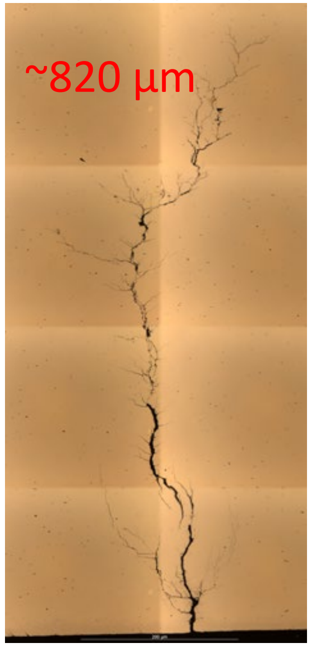
X-ray tomography

Identification of pits and cracks.

Metallographic cross-sections

Identification of pit and crack depths

Cross-section



All great techniques for corrosion analysis but they require small sections of the 3013 container which means the container has to be DE.

Can we perform this level of corrosion analysis without sectioning the 3013 container?

Why is it important to maintain the 3013 container <u>intact</u>?

Being able to do corrosion analysis in the ICCWR of intact 3013 containers allow us to study the progression of corrosion in these container over time.

Benefits:

1. Monitoring corrosion over extended period of time without interrupting the experiment
2. Observing changes in corrosion rate for better understanding the reaction kinetics
3. Measuring crack growth rate over time
4. Inspection of pits and cracks at the moment of formation

Attenuation coeffitients for thermal neutrons [cm^{-1}]

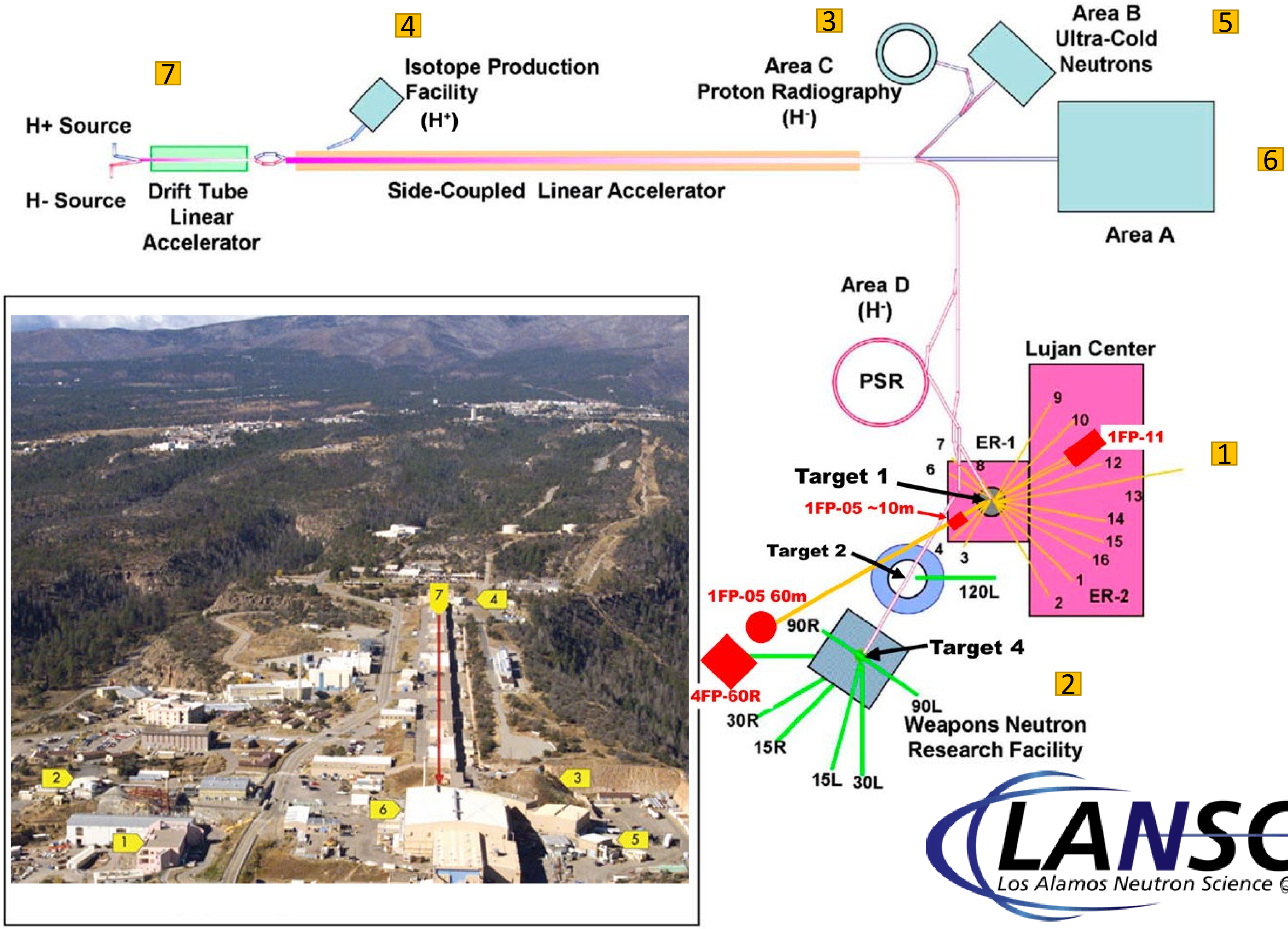
	1a	2a	3b	4b	5b	6b	7b	8			1b	2b	3a	4a	5a	6a	7a	0
	H 3.44																	He 0.02
	Li 3.30	Be 0.79											B 101.60	C 0.56	N 0.43	O 0.17	F 0.20	Ne 0.10
	Na 0.09	Mg 0.15											Al 0.10	Si 0.11	P 0.12	S 0.06	Cl 1.33	Ar 0.03
	K 0.06	Ca 0.08	Sc 2.00	Ti 0.60	V 0.72	Cr 0.54	Mn 1.21	Fe 1.19	Co 3.92	Ni 2.05	Cu 1.07	Zn 0.35	Ga 0.49	Ge 0.47	As 0.67	Se 0.73	Br 0.24	Kr 0.61
	Rb 0.08	Sr 0.14	Y 0.27	Zr 0.29	Nb 0.40	Mo 0.52	Tc 1.76	Ru 0.58	Rh 10.88	Pd 0.78	Ag 4.04	Cd 115.11	In 7.58	Sn 0.21	Sb 0.30	Te 0.25	I 0.23	Xe 0.43
	Cs 0.29	Ba 0.07	La 0.52	Hf 4.99	Ta 1.49	W 1.47	Re 6.85	Os 2.24	Ir 30.46	Pt 1.46	Au 6.23	Hg 16.21	Tl 0.47	Pb 0.38	Bi 0.27	Po	At	Rn
	Fr	Ra 0.34	Ac	Rf	Ha													
*Lanthanides		Ce 0.14	Pr 0.41	Nd 1.87	Pm 5.72	Sm 171.47	Eu 94.58	Gd 1479.04	Tb 0.93	Dy 32.42	Ho 2.25	Er 5.48	Tm 3.53	Yb 1.40	Lu 2.75			
**Actinides		Th 0.59	Pa 8.46	U 0.82	Np 9.80	Pu 50.20	Am 2.86	Cm	Bk	Cf	Es	Fm	Md	No	Lr			

Attenuation coeffitients for X-ray [cm^{-1}] (150kV)

	1a	2a	3b	4b	5b	6b	7b	8			1b	2b	3a	4a	5a	6a	7a	0
	H 0.02																	He 0.02
	Li 0.06	Be 0.22											B 0.28	C 0.27	N 0.11	O 0.16	F 0.14	Ne 0.17
	Na 0.13	Mg 0.24											Al 0.38	Si 0.33	P 0.25	S 0.30	Cl 0.23	Ar 0.20
	K 0.14	Ca 0.26	Sc 0.48	Ti 0.73	V 1.04	Cr 1.29	Mn 1.32	Fe 1.57	Co 1.78	Ni 1.96	Cu 1.97	Zn 1.64	Ga 1.42	Ge 1.33	As 1.50	Se 1.23	Br 0.90	Kr 0.73
	Rb 0.47	Sr 0.86	Y 1.61	Zr 2.47	Nb 3.43	Mo 4.29	Tc 5.06	Ru 5.71	Rh 6.08	Pd 6.13	Ag 5.67	Cd 4.84	In 4.31	Sn 3.98	Sb 4.28	Te 4.06	I 3.45	Xe 2.53
	Cs 1.42	Ba 2.73	La 5.04	Hf 19.70	Ta 25.47	W 30.49	Re 34.47	Os 37.92	Ir 39.01	Pt 38.61	Au 35.94	Hg 25.88	Tl 23.23	Pb 22.81	Bi 20.28	Po 20.22	At	Rn 9.77
	Fr	Ra 11.80	Ac 24.47	Rf	Ha													
*Lanthanides		Ce 5.79	Pr 6.23	Nd 6.46	Pm 7.33	Sm 7.68	Eu 5.66	Gd 8.69	Tb 9.46	Dy 10.17	Ho 10.91	Er 11.70	Tm 12.49	Yb 9.32	Lu 14.07			
**Actinides		Th 28.95	Pa 39.65	U 49.08	Np	Pu	Am	Cm	Bk	Vf	Es	Fm	Md	No	Lr			

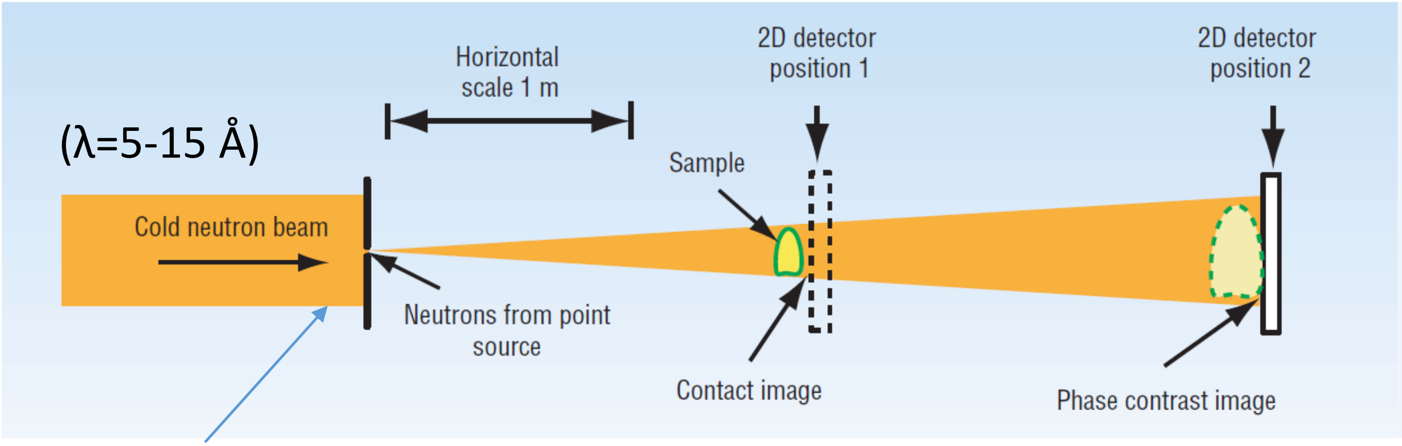
Why neutrons?

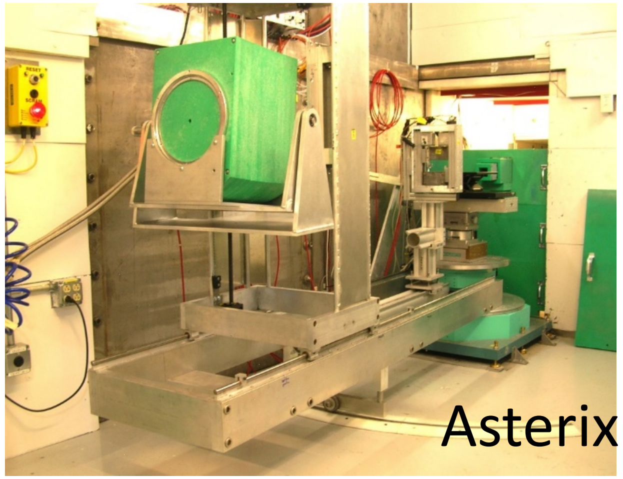
1. Higher resolution
2. Deeper penetration of material
3. Better contrast among elements with similar Z
4. Neutrons activate materials, so built-in infrastructure to mitigate radioactive hazards
5. Neutrons are isotope selective



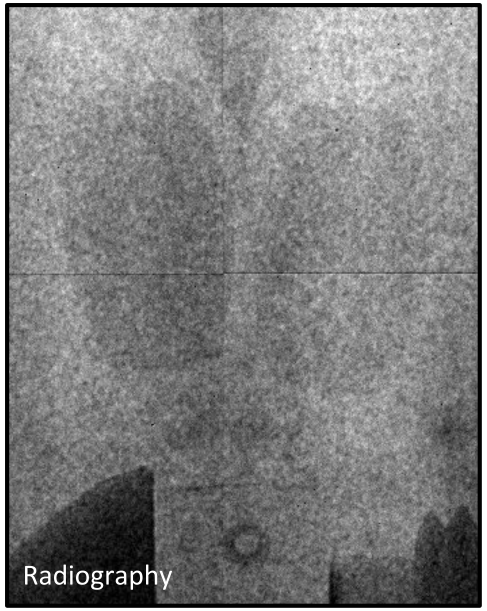
LANSCE
Los Alamos Neutron Science Center

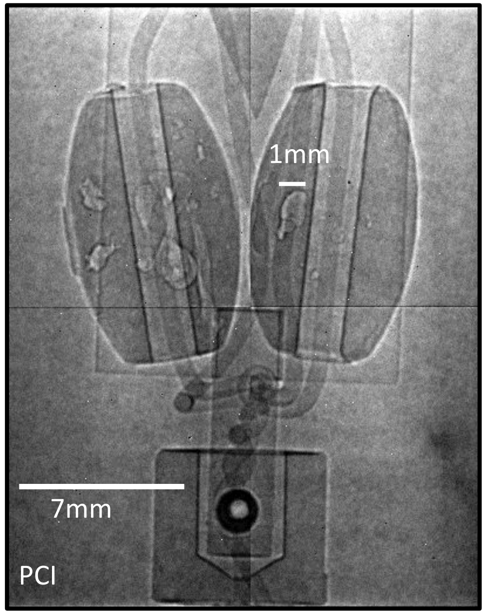
Cold neutron phase contrast imaging (PCI) at LANSCE: in-line propagation method (resolution >50 μm)





Needs high spatial coherence, results in low flux and long count times (~24hrs)



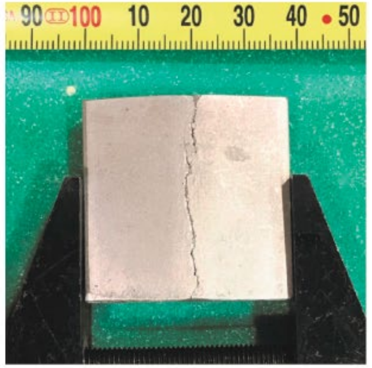


PCI of Pb fishing weights:

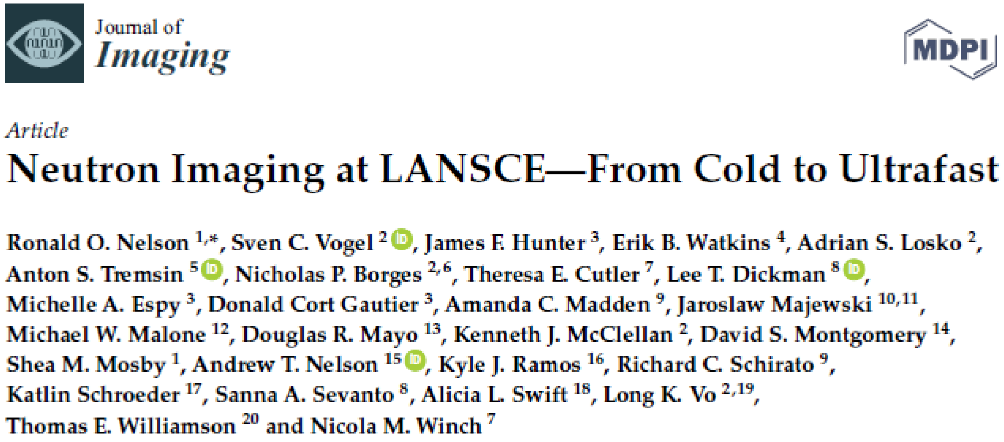
Due to the low absorption contrast, a radiographs contain little information (center).

Phase contrast imaging (~50μm resolution) was used to enhance the visibility of interfaces and interior defects in the Pb (right).

More capabilities of neutron PCI



Photograph of
cracked 50x50 mm Al plate
Crack width: 150 μm

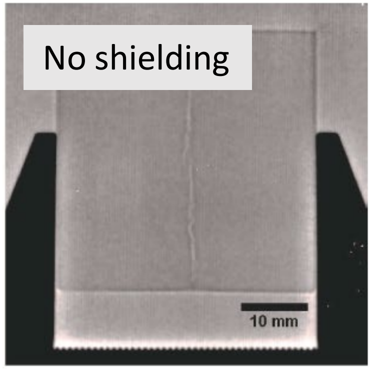

Journal of *Imaging*

MDPI

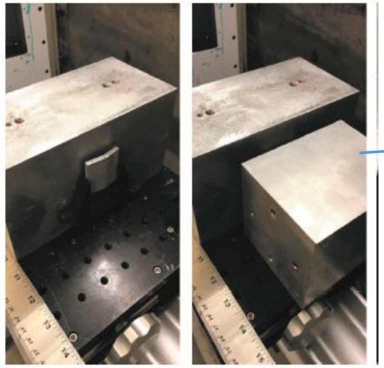
Article

Neutron Imaging at LANSCE—From Cold to Ultrafast

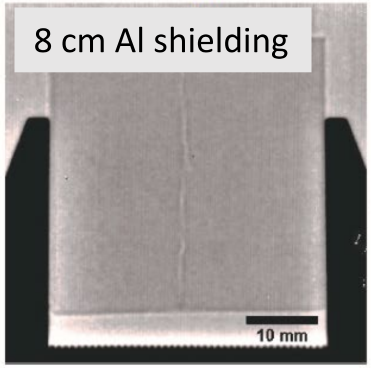
Ronald O. Nelson [1,*], Sven C. Vogel [2], James F. Hunter [3], Erik B. Watkins [4], Adrian S. Losko [2], Anton S. Tremsin [5], Nicholas P. Borges [2,6], Theresa E. Cutler [7], Lee T. Dickman [8], Michelle A. Espy [3], Donald Cort Gautier [3], Amanda C. Madden [9], Jaroslaw Majewski [10,11], Michael W. Malone [12], Douglas R. Mayo [13], Kenneth J. McClellan [2], David S. Montgomery [14], Shea M. Mosby [1], Andrew T. Nelson [15], Kyle J. Ramos [16], Richard C. Schirato [9], Katlin Schroeder [17], Sanna A. Sevanto [8], Alicia L. Swift [18], Long K. Vo [2,19], Thomas E. Williamson [20] and Nicola M. Winch [7]



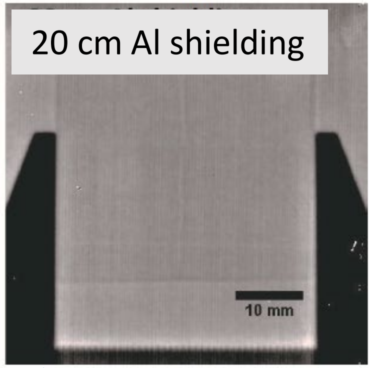
Phase contrast
image using the
propagation method,
10 minutes exposure



photos of the cracked plate between
20 cm of Al shielding

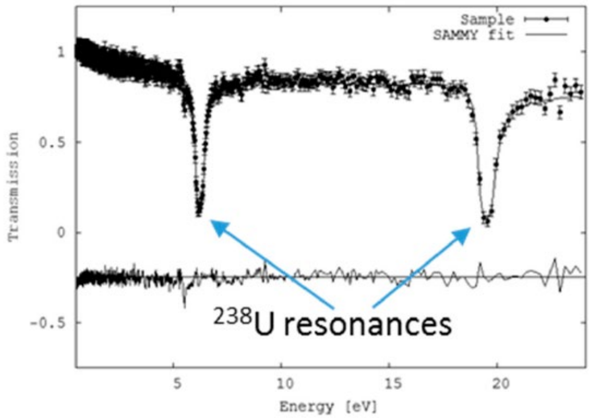


PCI of the plate
inside 8 cm of Al
shielding, 20 minutes
exposure

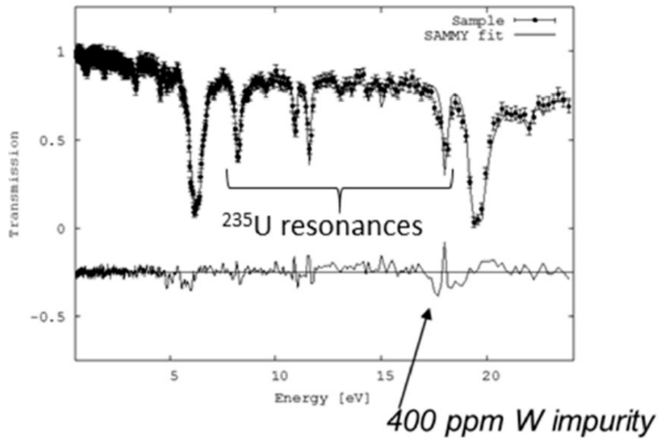


PCI of the plate inside
20 cm of Al shielding,
25 minutes exposure

More capabilities of neutron imaging: Isotope separation



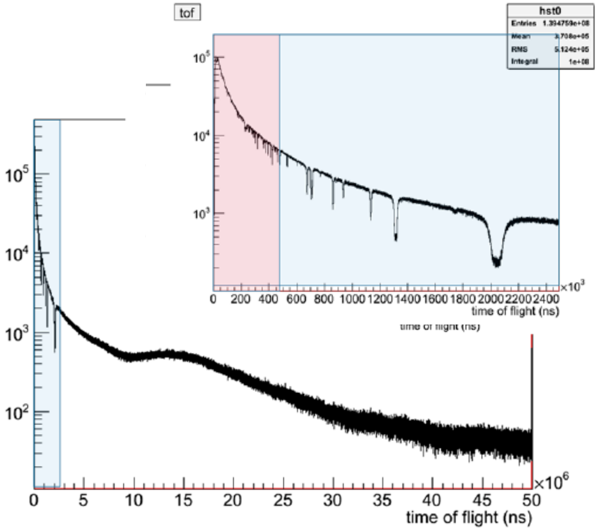
0.2% ^{235}U U_3Si_5



8.8% ^{235}U U_3Si_5

Examples of transmission data analysis for two fuel samples with different enrichment

Sample data collected with a liquid scintillator detector.

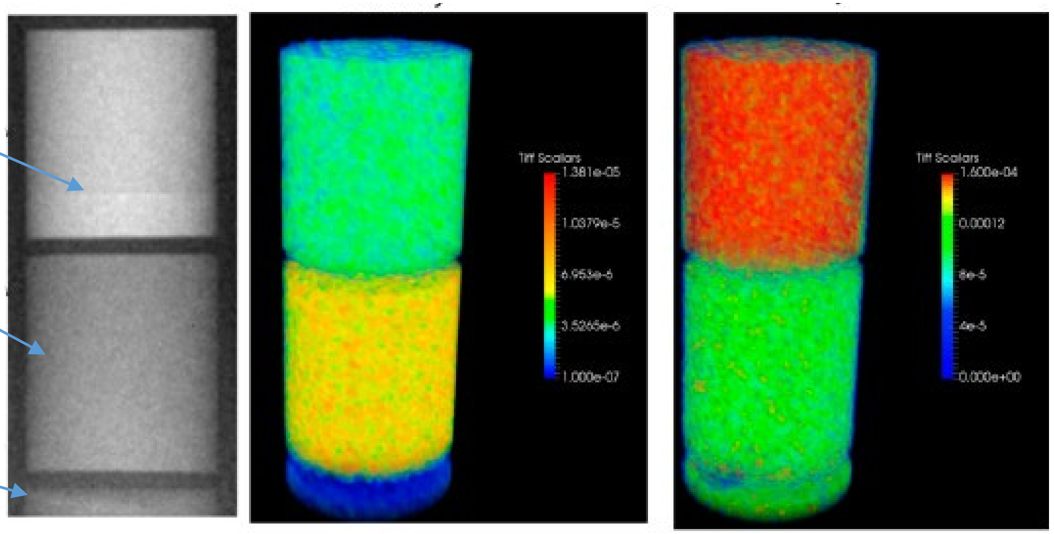


UN/U_3Si_5 and U_3Si_5 fuel pellets with different enrichment levels

U-235 0.34 g/cc
in U-238 12.43 g/cc

U-235 0.61g/cc
in U-238 6.91 g/cc

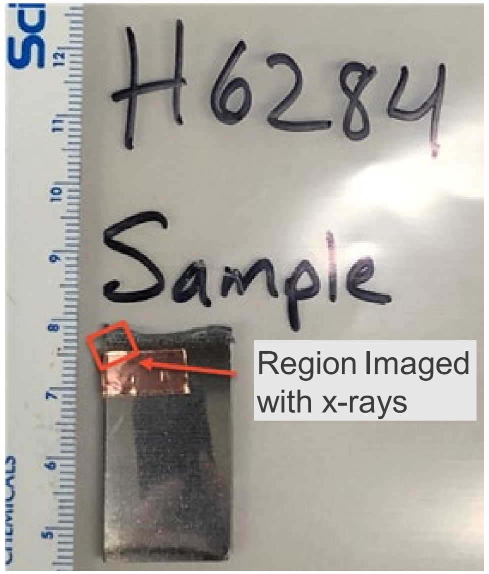
U-235 0.02g/cc
in U-238 7.56 g/cc

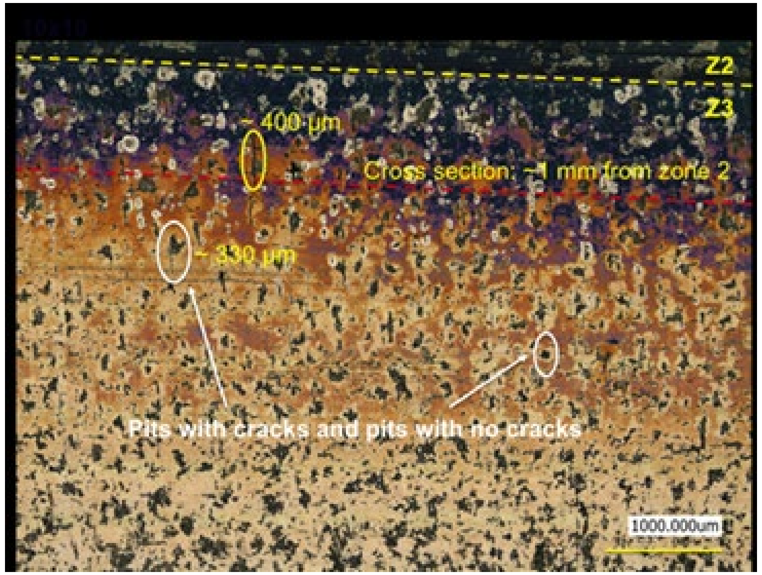


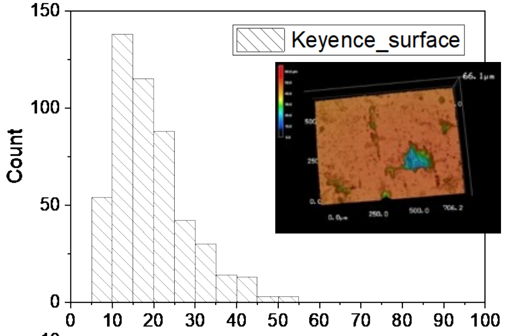
tomography

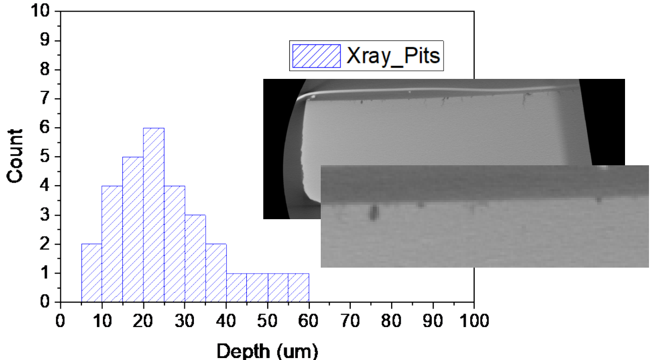
Neutron computed tomography

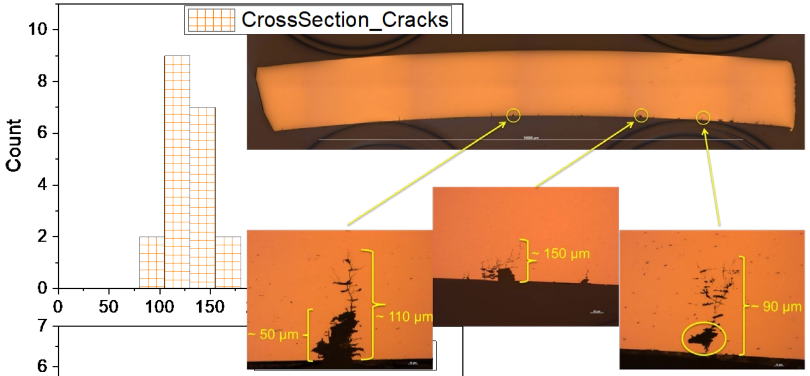
Can we perform this level of corrosion analysis without sectioning the 3013 container?

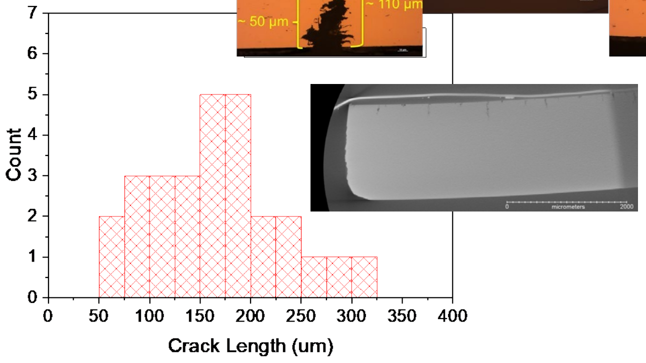






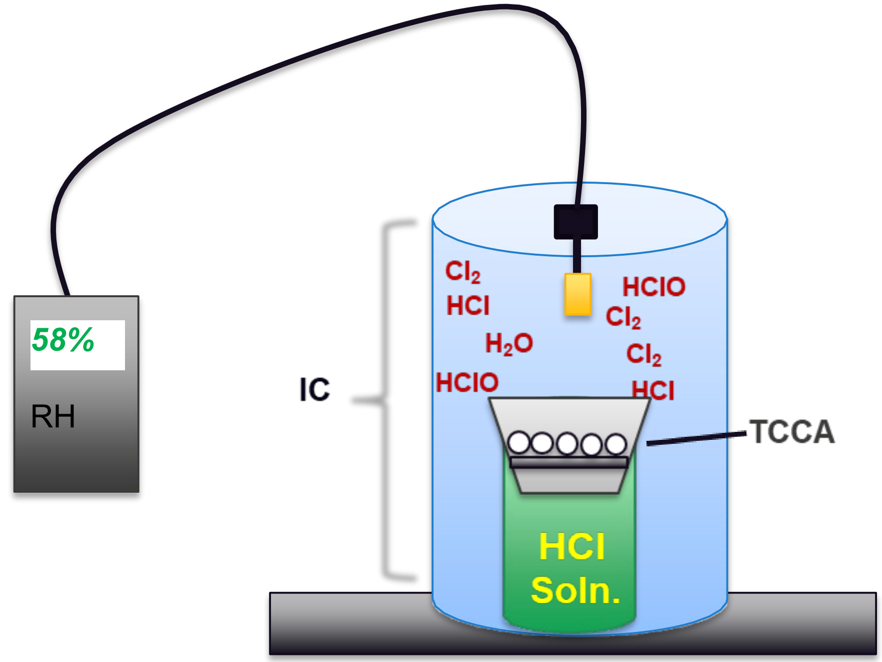






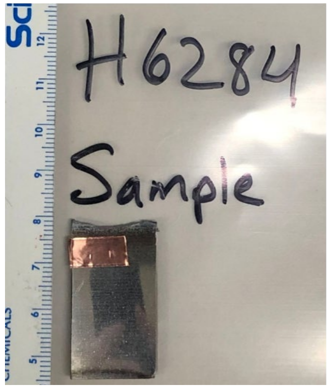
- *Exposure to corrosive conditions for 109 days*

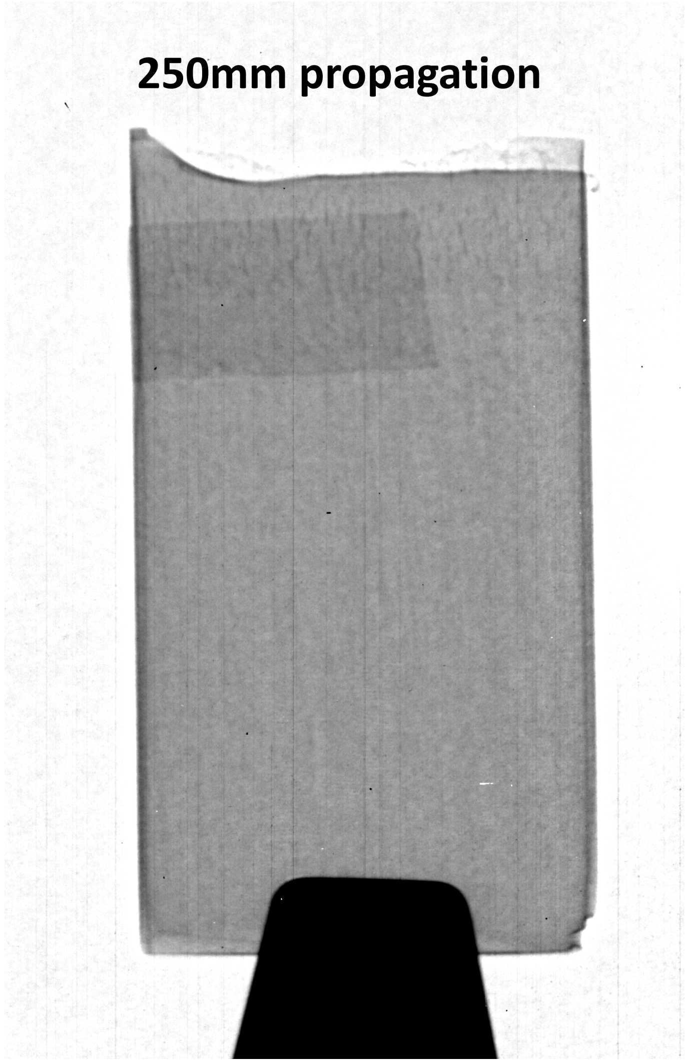
- *4.5 mL of 5.4 M HCl*
- *2.39 g of TCCA*

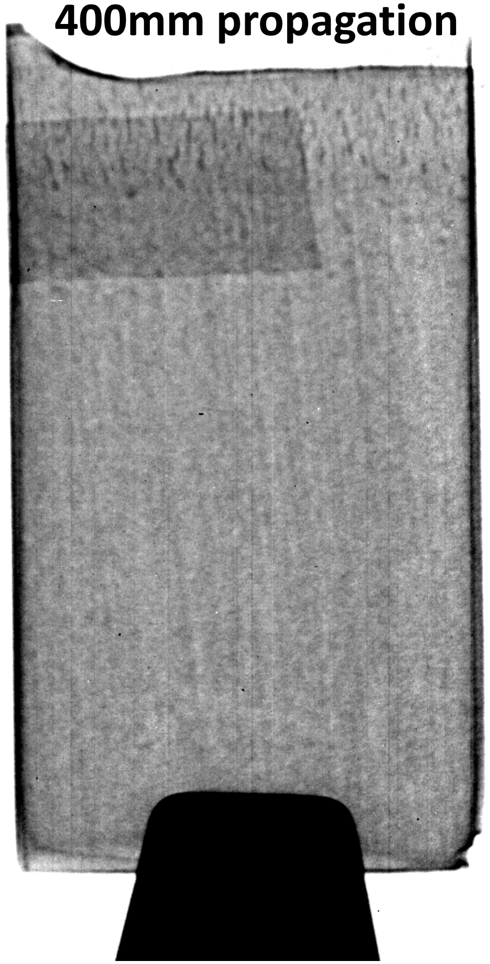


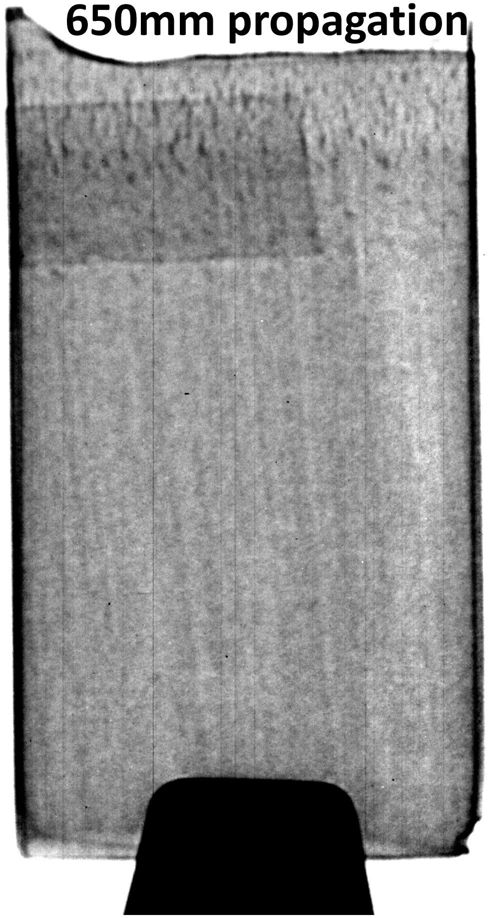
In-line propagation neutron imaging of 3013 container fragment

Transmission based neutron radiography does not detect any inhomogeneity in the fragment.
Using in-line propagation imaging, ~100-500 micron features appear at the top of the sample.
Increasing propagation distance increases contrast but decreases resolution.









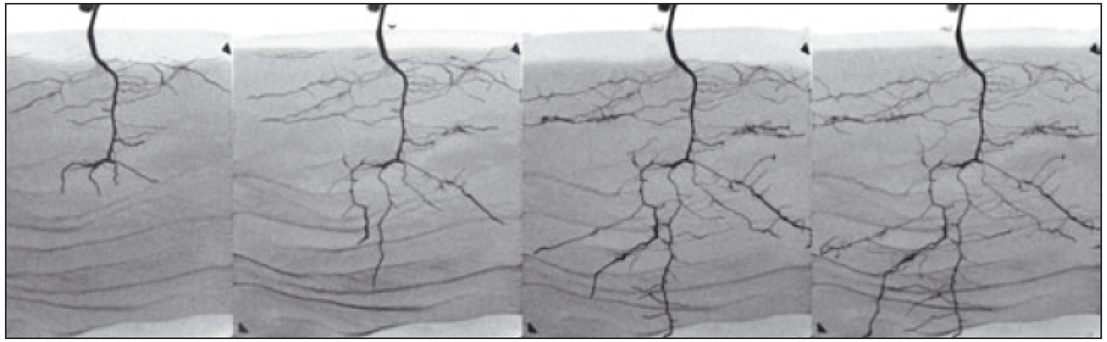
The H6284 piece of IC was imaged at following conditions:

Neutron wavelengths: 4-13 Angstroms
Source: 2mm diameter pinhole
Source-to-detector distance: 5.75 m
Camera: ATIK 490 EX CCD
Scintillator: 10x10cm, 35 micron resolution, ZnS:Cu/6LiF (RCTritec)

Proof of Concept experiments

Obtain neutron PC Images of ICCWR samples and Compare with LCM results:

- 1/8 ICCWR exposed to HCl gas and 1/8 ICCWR exposed to Cl_2 gas
- ICCWR of pristine full scale container to establish baseline and neutrons transmission



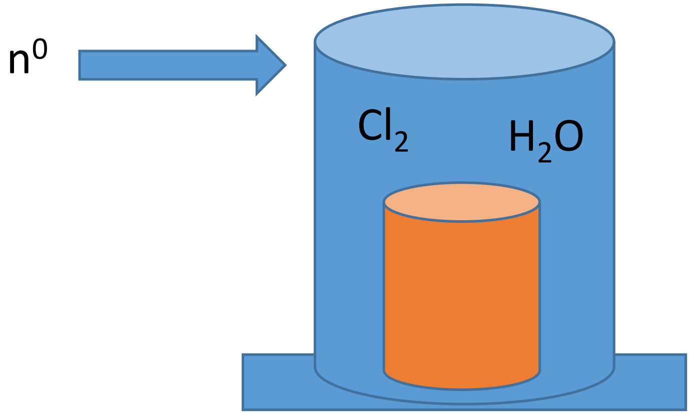
Examination of root growth of lupin over 4 weeks as an example of an in situ, continuous experiments

Achievements: whole piece of the sample imaged; corrosion features identified also sub-surface

Experiments *in situ*

Based on proof of concept results, neutron images of ICCWR could be collected on a single 3013 inner container being exposed to Cl_2 gas at 50% RH:

- Baseline measurement on a pristine 3013 inner container
- ICCWR area of an intact IC exposed to HCl gas
- ICCWR area of an intact IC exposed to Cl_2 gas



Further plans: Checking whether the corrosion effects continue developing under conditions as observed in DE 3013 (lower RH)

Concluding remarks

1. Goal for Non-Destructive Analysis: Ability to study crack growth over time and evaluate impact of various parameters on crack growth
2. Potential of Neutron Imaging for corrosion analysis of 3013 containers
3. Neutron source available
4. No reports of in situ corrosion monitoring inside objects
5. DOE and NNSA mission relevance: defining safety envelope of storage of the plutonium oxide inventory that contains chloride salts
6. Challenges: new technique under development, relying on the accelerator cycle (6 months not operating)